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Pedon-scale silicate weathering: comparison of the PROFILE model and the depletion method at 16 forest sites in Sweden

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Abstract

Weathering of soil minerals is important for the recovery from acidification and for the sustainability of forestry. However, there is still substantial uncertainty about its absolute rate. This study presents a harmonized comparison of field weathering rates estimated with the mechanistic model PROFILE and the depletion method for 16 intensively sampled soil profiles across Sweden representing different site conditions. In general, a correspondence in total weathering rates was found between the two methods except in rare cases where either method yielded deviating results. The weathering rate was higher according to the depletion method than according to PROFILE for Mg, while PROFILE produced higher weathering rates for the other base cations. The Spearman rank correlation (ρ) between the two methods indicated significant correlation for Ca ($\rho=0.44$, $p=0.04$) and non-significant correlation for Mg ($\rho=0.51$, $p=0.09$), Na ($\rho=0.25$, $p=0.34$), K ($\rho=0.07$, $p=0.80$), and the sum of the base cations ($\rho=0.11$, $p=0.67$). The variation in weathering rates with depth showed opposite gradients in the upper 50 cm, which reflects the conceptual differences between the methods. This study shows the potential of using multiple methods to identify a probable weathering rate, if harmonized input data is used. Furthermore, it highlights the importance of making comparisons for individual elements in order to interpret differences between methods. Regardless of the method used, weathering rates were below or at the same level as the losses caused by whole-tree harvesting, particularly in southern Sweden, indicating a risk of negative effects on soils and waters.

1. Introduction

Chemical weathering is important for the nutrient sustainability of forests, the neutralization of acidifying compounds and the quality of water transported from upland soils to surface water systems downstream. The historical deposition of acid compounds, which culminated around 1980 (Schöpp et al., 2003), caused substantial leaching of base cations from soils to waters, leading to base cation depletion and acidification of soils. A reduction of acid deposition to levels below the total of base cation weathering and base cation deposition is a prerequisite for the recovery of soils from acidification. However, recovery will also depend on the export of nutrients from the forest ecosystem through harvesting, which has increased recently owing to the increased focus on using harvest residues for bioenergy, e.g. whole-tree harvesting. While active measures, such as ash recycling, are recommended in e.g. Sweden (Swedish Forest Agency, 2008) a basis for such activities is an assessment of the mass balance of input and output of mineral nutrients in the ecosystem. Thus, there is a demand for robust estimates of the release rate of mineral nutrients by weathering for predictions of future recovery from acidification and for optimizing forest management policies.

Proposed methods for determining weathering rates include the mass balance approach using experimental data (Bain et al., 1994), the mass balance approach using catchment modelling (Cosby et al., 2001), the depletion method using an immobile element as an internal standard (Brimhall & Dietrich, 1987; Olsson & Melkerud, 1989), and the process oriented model PROFILE where weathering rates are estimated for soil pedons (Sverdrup & Warfvinge, 1993). The various methods are conceptually different and are intended for different spatial scales; from the soil pedon at site level to the entire soil deposit at the catchment level. Furthermore, the estimates consider different time perspectives; from long-term historical averages to present day weathering and steady state weathering.

Several attempts have been made to quantify weathering rate uncertainties through an ensemble approach (Kolka et al., 1996; Starr et al. 1998; Sverdrup et al., 1998; Whitfield et al. 2006; Koseva et al., 2010; Klaminder et al., 2011; Futter et al., 2012). Sverdrup et al. (1998) compared rates from six different approaches in two catchments in Gårdsjön, Sweden. The variation in base cation weathering rates (Ca, Mg, Na and K) was 30-39 and 47-81 meq m^{-2} y^{-1} in the two catchments respectively, indicating relatively low uncertainties. On the other hand, Klaminder et al. (2011) demonstrated a wide span for Ca and K weathering rates, 7-150 meq m^{-2} y^{-1} and 2-32 meq m^{-2} y^{-1} respectively, at a site in northern Sweden where ten studies were compared, and concluded that estimated weathering rates were too uncertain to be used in assessments of sustainable harvesting. They revised their conclusions slightly in Futter et al. (2012), where they pointed out that much of the variation was related to input data and known conceptual differences between the methods, for example that different estimations were valid for different soil depths. They concluded that at least three independent estimates should be used when making management decisions. This is in line with the conclusions by Whitfield et al. (2006) who compared weathering estimates from five approaches in five catchments in Canada. The three soil profile-based approaches gave similar results, with low weathering rates ranging from 3 to 13 meq m^{-2} y^{-1}. However, the two catchment-based methods gave one order of magnitude higher rates. Koseva et al. (2010) compared PROFILE weathering estimated with catchment mass balance calculations for 19 sites. The PROFILE weathering rates were in most cases within the range of the catchment mass balance weathering rates, but PROFILE weathering rates were generally lower, as expected when comparing soil pedon weathering rates with catchment weathering rates. Although they did not present any direct uncertainty measures based on this, they used it to demonstrate the reliability of PROFILE. A critical aspect when comparing weathering estimates is the quality of the input data, boundary conditions and assumptions made, e.g. the maximum soil depth considered and corrections for coarse fragments and organic matter. The stone and boulder content may amount to $>50\%_{vol}$ in many forest soils (Stendahl et al., 2009) and the way it is treated will have a strong influence on estimated pedon-scale weathering rates. It is crucial that comparisons of weathering estimation methods are harmonized with regard to assumptions and input data, and they should be carefully evaluated when published results from different studies are synthesized.

In this study we estimated the base cation weathering at the pedon-scale by the depletion method and PROFILE for 16 intensively sampled soil profiles representing a wide range of soil conditions in Sweden. Both methods have been frequently used to estimate weathering rates on local, regional and national scale for assessments of sustainable harvesting (Olsson et al., 1993; Sverdrup & Rosén, 1998; Akselsson et al., 2007). The depletion method quantifies the loss of mobile elements since the parent material was deposited, whereas PROFILE estimates the release of elements due to the dissolution of soil minerals at steady state. Hence, the two methods consider very different time perspectives. Despite the different time perspectives of the methods the comparison was justified by the relatively young age of the profiles (10 000 – 16 000 years). The methods were applied in a harmonized way using input data from the same forest sites and the same pits, as well as using the same assumptions. Furthermore, the estimated weathering rates were compared with estimated base cation losses at whole-tree harvesting on a selection of the sites, in the same way as in e.g. Olsson et al. (1993) and Klaminder et al. (2011). This simplified mass balance calculation puts the difference in weathering rates between the two methods in a sustainability perspective. The objectives of this study were to: (i) compare how the two methods ranked the sites with regards to Ca, Mg, K, and Na weathering, (ii) compare how weathering intensity vary with depth in the uppermost soil profile for the two methods, (iii) investigate the causes for deviating results between the two methods, and (iv) put the results in a sustainability perspective by comparing the estimated weathering rates with base cation losses associated with whole-tree harvesting.

The comparison can shed light on how the total weathering rate and weathering intensity within the soil profile evolve over time and can be seen as a robustness test of the two methods.

2. Material and Methods

2.1. Sites and soil profile data

The 16 sites, which are part of the NORDSOIL database (Raulund-Rasmussen & Callesen, 1999), are located at latitudes 56-68 °N across Sweden on podzolised glacial till without stratigraphic layering (Fig. 1, Table 1). All sites are located above the highest Quaternary shoreline and it was assumed that no redistribution of soil material has occurred since deglaciation. The parent material is of granitic composition with varying mineralogical composition. Among the sites 12 were used previously in weathering studies by Olsson & Melkerud (1989, 1991, 2000) and Olsson et al. (1993). For each site, a profile was used that had complete soil chemical and physical data necessary to apply the two methods. Sampling was made at 10 cm depth intervals from the top of the mineral soil to the maximum mineral soil depth (42-263 cm, Table 1). Volumetric samples were taken by a core sampler for each 10 cm layer in the A, B and uppermost C horizon and bulk density was determined after drying and weighing. Grain size distribution data was determined for 10 cm layers, although it was missing for the A horizons and ca 25% of the other layers. In those cases the grain size distribution was estimated based on the layers above and below. For each site the stone content was estimated based on the soil texture class to 10-40%$_{vol}$. An analysis of the total geochemical content of the bulk soil material <2 mm was made for the samples from each 10 cm layer throughout the profile by extracting a representative subsample (20 g) that was homogenized by dry grinding in an agate mortar after which 0.125 g was fused with 0.375 g lithium metaborate ($LiBO_2$) at 1000 ^{o}C and dissolved in nitric acid. The content of major elements and trace elements was quantified by plasma emission spectrometry analysis (ICP-AES). The analytical data were normalized with respect to dry matter weight.

2.2. The depletion method

The basic principle of the method is the long-term depletion of mobile (weatherable) elements and the concurrent enrichment of immobile (inert) elements in weathered soil layers during soil development. The concentration of the mobile and the immobile element in a weathered and an unweathered soil layer are utilized to estimate the loss of the mobile element in the weathered layer. The idea to use immobile elements to estimate element losses was first proposed by Marshall & Haseman (1942) and the theoretical framework was formalized by Brimhall & Dietrich (1987) and Brimhall et al. (1991). Their framework focuses on the relative mass loss (or gain) of elements from layers in the original soil pedon and they define the fractional mass loss as:

$$\tau_{j,w} = \frac{C_{j,w} C_{i,p}}{C_{j,p} C_{i,w}} - 1 \qquad (1)$$

where C denote concentrations (%), i denotes the immobile element, j denotes the mobile element, w denotes a weathered horizon, and p denotes the unweathered parent material. Values of $\tau < 0$ indicates net mass loss, $\tau > 0$ net mass gain, and $\tau = 0$ no change.

The weathered amount of the mobile element from a soil layer (g m^{-2}) can be expressed based on the same principles (Olsson & Melkerud 1989):

$$W_{j,w} = \frac{d_w \rho_w}{100} \left(C_{j,p} \left(\frac{C_{i,w}}{C_{i,p}} \right) - C_{j,w} \right) \qquad (2)$$

where d is layer depth (m), and ρ is bulk density (g m^{-3}).

Zirconium, which is predominantly found in the weathering resistant mineral zircon ($ZrSiO_4$), was used in this study as the immobile element, since its redistribution within the profile is negligible (Hodson, 2002). The depletion method assumes that there is no weathering of the immobile element, that there is no weathering below a certain soil depth, and that the soil profile was developed from homogenous regolith. One critical aspect of the method is to define a reference soil layer to represent the unweathered parent material.

In this study we used equation (2) to estimate the loss of mobile base cations. The reference soil layer used (Table 2) was the uppermost layer that met the following criteria: (1) it was located in the soil C horizon, (2) the ratio between the mobile element and Zr was stable below this layer, and (3) the Zr gradient was stable below this layer. The long-term weathering rate was calculated by dividing the total weathered amount by the soil age, i.e. the age since deglaciation given by the National Atlas of Sweden (Fredén, 2009; Table 1).

2.3. The PROFILE model and input data preparation

PROFILE is a steady state soil chemistry model, originally developed to calculate the effect of acid rain on soil chemistry (Sverdrup & Warfvinge, 1993). PROFILE includes process oriented descriptions for chemical weathering of minerals, leaching and accumulation of dissolved chemical components and solution equilibrium reactions. The weathering module is a central part of the model. Weathering rates are calculated using transition state theory, based on the geochemical properties of the soil system, climate, deposition and tree uptake. Soil physical properties, such as soil mineralogy, soil moisture content, bulk density and exposed mineral surface area, have been identified as the input parameters with the highest influence on the output in several studies (Jönsson et al., 1995; Hodson et al., 1996; Zak et al., 1997).

In PROFILE, the soil is divided into soil layers with different properties, preferably based on the naturally occurring soil horizons. Soil chemistry and weathering rates are calculated for each layer, and the soil chemistry of each layer is affected by the layer above, through vertical water flow.

The estimated soil chemistry and weathering rates describe the situation at steady state, i.e. the soil chemistry and weathering rates that will eventually evolve, with the deposition, tree uptake, climate conditions and soil properties that are given as input to the model (Sverdrup & Warfvinge, 1993). Thus, the model ignores development over time, e.g. changes in soil properties due to weathering.

Detailed soil data was available for the chosen profiles from the NORDSOIL database and was used in the input data preparation for the PROFILE model runs. Total elemental content for the different soil layers was used, along with information about predominant minerals and their compositions compiled in previous studies (Warfvinge & Sverdrup, 1995; Akselsson et al., 2004), for calculating soil mineralogy with the A2M model (Posch & Kurz, 2007). The modeled mineralogical composition for the sites is given in Table 3. The specific surface area of minerals, i.e. the weatherable area, was calculated using an empirical algorithm (Warfvinge & Sverdrup, 1995; Holmqvist et al., 2002) using data on the grain size distribution and bulk densities for each layer (data from 50 cm given in Table 1). Data on gravel content were only available for a few of the sites, for the rest of the sites the gravel content was estimated based on grain size distribution (Table 1) and sieving curves (SGU, 1994). The estimated specific surface area was corrected for the stone volume in the soil by subtracting the stone volume from the soil volume, assuming that the weathering from stones is negligible due to the low specific surface area as compared with fine soil. Soil moisture class was given for each site according to a classification system with seven classes (Table 1). Soil moisture was allocated to the soil moisture classes, according to methods in Warfvinge & Sverdrup (1995). Concentration of DOC, CO_2 pressure and Al solubility constants for different layers were derived from Martinson et al. (2003).

Information on tree species and mean annual production from the NORDSOIL database (Table 1) was used for estimating net uptake according to methods described in Akselsson et al. (2007), using base cation concentrations in stem biomass compiled in Akselsson (2005). Mean annual temperature and precipitation was derived from the NORDSOIL database (Table 1), whereas runoff was derived from national maps from the Swedish Meteorological and Hydrological Institute (SMHI) (Raab & Vedin, 1995). Deposition values were derived from modeled deposition values for 1998 by the MATCH model (Langner et al., 1996).

2.4. Comparison with base cation losses at whole-tree harvesting

The base cation losses were estimated for stem (log) only and whole-tree harvesting for the eight spruce sites. The calculations were based on the mean annual stem biomass production for each site (Table 1) and the amount of stems, branches and needles was estimated using biomass expansion functions (Marklund, 1988), which are based on empirical relationships between the diameter at breast height and empirical biomass data for a large number of trees in Sweden. The amount of stems, branches and needles was multiplied with concentrations of Ca, Mg, K and Na in the different tree compartments compiled in Akselsson (2005). Stem harvesting was defined as removal of the stem only, whereas whole-tree harvesting was defined as the harvest of the stem together with 60% of the branches in thinnings and in final felling (Swedish Forest Agency, 2008). Furthermore, it was assumed that 75% of the needles on 60 % of the branches, i.e. in total 45% of the needles, were removed. The methodology is described in more detail in Akselsson et al. (2007).

3. Results

The comparison of the total base cation weathering rates in the upper 50 cm of the mineral soil showed significantly (p=0.006, t-test) higher rates for PROFILE (with an interval of 12-135 meq m^{-2} yr^{-1} and a median of 39 meq m^{-2} yr^{-1}) than for the depletion method (with a corresponding interval of 8-41 meq m^{-2} yr^{-1} and a median of 19 meq m^{-2} yr^{-1}). There were substantial deviations in estimated weathering rates for three sites. The rate estimated by the PROFILE model was extremely high in Nunasvaara for Ca (35 meq m^{-2} yr^{-1}), Mg (40 meq m^{-2} yr^{-1}) and Na (51 meq m^{-2} yr^{-1}); 5 times higher than the rates derived from the depletion method. In Målaskog the PROFILE-modeled Ca weathering rate was also high (31 meq m^{-2} yr^{-1}) compared to the depletion method (6.6 meq m^{-2} yr^{-1}). In Vålberget it was the other way around, with high rates of Ca (21 meq m^{-2} yr^{-1}) and Mg (13 meq m^{-2} yr^{-1}) weathering from the depletion method compared to PROFILE (4.8 and 2.3 meq m^{-2} yr^{-1} for Ca and Mg respectively).

For Ca and Mg there was correlation between the results from the two methods (Fig. 2). The weathering of Mg was generally higher according to the depletion method, whereas it was the other way around for Ca. For K and Na there was weak or no correlation between the two methods. PROFILE gave generally higher weathering rates for Na, whereas there was no such difference for K. The Spearman rank correlation between the depletion method and PROFILE (Table 4) indicated significant correlation for Ca (ρ=0.44, p=0.04), while it was non-significant for Mg (ρ=0.51, p=0.09), Na (ρ=0.25, p=0.34) and K (ρ=0.07, p=0.80). For the sum of the base cations the correlation was non-significant (ρ=0.11, p=0.67). The correlation coefficients for Ca and Mg were strongly influenced by the three extreme sites where either method gave highly deviating results. Disregarding these sites resulted in highly significant correlations for Ca (ρ=0.71, p=0.007) and Mg (ρ=0.82, p=0.001). For Ca and Na weathering estimated with the same method there was a highly significant correlation; ρ=0.80 (p<0.001) for PROFILE and ρ=0.73 (p=0.001) for the depletion method.

The average mass loss estimated by the depletion method (Fig. 3) increased towards the surface in the investigated profiles, but the gradients varied for the different base cations. The largest average mass loss in the uppermost mineral soil was found for Mg (66%), followed by Ca (43%), Na (38%) and K (29%) as compared to the reference soil layers. The gradient in weathering rate with depth was the opposite for the two methods (Fig. 4). The depletion method, reflecting the average rates since the last glaciation, showed high rates in the upper layer, and decreasing rates with depth, reaching ca 12 meq m^{-3} yr^{-1} at the 50 cm level. The PROFILE results showed, on the other hand, an increasing weathering rate with depth, reaching ca 110 meq m^{-3} yr^{-1} at the 50 cm level.

The calculation of harvest losses of base cations showed that harvesting of branches and needles almost doubled the losses compared with stem harvest only (Fig. 5). The comparison between harvest losses and weathering rates showed that the losses at whole-tree harvesting were of the same order of magnitude as PROFILE modeled weathering rates for the three northern sites, whereas the weathering rates from the depletion method were about half the size. For the southern sites, the harvest losses at whole-tree harvesting were substantially higher than the weathering rates from either approach.

4. Discussion

4.1. Comparison between results and concepts

The weathering rates estimated by the two methods agreed in some respects, but not in others and the observed differences may be due to a number of factors. The depletion method gives the average weathering rate since the deglaciation, but the historical development of the weathering in the soil is not fully known. Steady state weathering, on the other hand, is a theoretical concept used in PROFILE, which is based on steady state calculations where the input data are kept constant at today’s levels, and the steady state soil chemistry and weathering are estimated under the assumption that the present minerals are not depleted. The difference in time perspective for the two methods makes it crucial to consider the historical development of the weathering rate in the interpretation of the results. The influence of soil age on the weathering rate has been investigated in chronosequence studies (Bain et al., 1993; Taylor & Blum, 1995; White et al., 1996; Hodson & Langan, 1999) and by modelling (Warfvinge et al., 1995). The results show high initial weathering rates followed by a decline, which is most pronounced in the first centuries following soil exposure, but the results varies greatly between sites. The decline is caused by the depletion of easily weathered minerals and decrease in reactive mineral surface area (Taylor & Blum, 1995). Attempts have been made to correct the long-term weathering rate to today’s levels (Taylor & Blum, 1995) using a power-law function fitted to depletion weathering rates from chronosequence data. These results together with simulations of historical weathering rates (Warfvinge et al., 1995) indicate that the ratio between today's weathering rate and the long-term historical weathering rate is approximately 0.3-0.5.

Based on this, the depletion method was expected to give higher values than the PROFILE model. For Mg our results agreed with the theory; the depletion method gave higher weathering rates than PROFILE. However, for Ca, Na and K the PROFILE model gave higher values than the depletion method, although for K there was large non-systematic variation between the two approaches for many sites. Thus, the results for Ca, Na and K contradict the theory about decreasing weathering rates over time. A contributing factor to our observed differences between PROFILE and the depletion method is that the parent material for our profiles is relatively poor in easily weathered minerals that may give rise to such extreme initial weathering rates reported elsewhere (Bain et al., 1993; Taylor & Blum, 1995). Furthermore, the soil profiles were developed in glacial till that partly consists of soil material that was newly formed during the Weichselian glaciation and partly includes redistributed old till deposits from previous glaciations. Consequently, the parent material in the investigated profiles was not completely unweathered at the time of deglaciation. The relatively elevated acid deposition at present could also contribute to increased weathering (April et al. 1986), as well as the increased forest growth and harvest levels during the last century, leading to increased acidification and removal of weathering products. It is, however, difficult to design experiments aiming at quantifying these effects. A sensitivity analysis of PROFILE (Hodson et al., 1996) showed that increased sulphur deposition led to increased or unchanged weathering rates for most minerals, but the effect was generally relatively small.

The correlation analysis for Ca and Mg showed that the coefficients were negatively influenced by three sites where one of the two methods yielded highly deviating results. As discussed below, there are good reasons to believe that these deviating results were caused by conditions that caused either model to fail. Hence, the result indicates significant correlations for Ca and Mg weathering when conditions are suitable for both methods. For K and Na the correlation was weak and it could be hypothesized that K and Na weathering in PROFILE was more sensitive to variation in other input data than mineralogy than for Ca and Mg. This means that uncertainties in other input data would have a greater impact on the results. For K, this is to some extent supported by Hodson et al. (1996), who identified K-feldspar, one of the main sources of K, as a mineral particularly sensitive to input variations. To be able to fully explain the difference, even more in-depth studies are required, including an accurate determination of the actual minerals present. The Ca and Na weathering rates estimated with the same method were significantly correlated for both methods. This indicates a proportional release of Ca and Na that could be explained by the occurrence of both elements in the same minerals e.g. the plagioclases, which are rather weatherable and very common in the investigated area. The correlation study showed very different results for the different base cations, which highlights the importance to compare individual base cations rather than merely the sum of them.

Uncertainties related to the specific surface area are generally one of the main sources of uncertainties to the mineral weathering rates estimated with PROFILE. This can be due to lack of detailed data on grain size distribution, and due to the generalized equations transforming grain size distribution to specific surface area. Hodson et al. (1998) questioned the equation for estimating specific surface area from grain size distributions, based on a study in Scotland. However, since the equation was developed based on data from Sweden, the applicability can be expected to be higher in Sweden. Hodson and Langan (1999) highlighted the fact that changes over time of specific surface area and mineral reactivity are not taken into account in the PROFILE model, leading to uncertainties in the estimated weathering rates. The uncertainties introduced by the data on specific surface area can be reduced by using detailed input data on grain size distribution and densities for several layers, as was done in this study. However, although detailed data is used, the uncertainties will be substantial, due to the problems discussed above.

The depletion method showed very large mass loss of base cations in the upper part of the soil profile considering the relatively young age of the investigated soils (ca 10 000 – 16 000 years). Particularly the loss of Mg was considerable. It has been recognized that Mg is one of the most mobile elements in similar soils (Bain et al., 1993; Lång, 1995; Land et al., 1999; Olsson & Melkerud, 2000). In Land et al. (1999) they found a depletion level of 70% for Mg in the E horizon of Podzol soils in northern Sweden. The Mg loss is probably associated with depletion of easily weathered minerals such as chlorite and hornblende, which are important Mg-bearing minerals in the soils of the Svecofennian granitic bedrock and important for forest soil productivity (Stendahl et al., 2002). The high depletion levels for all base cations indicate a general loss of easily weathered minerals.

Important drivers for the weathering include processes at the soil surface such as biological production and deposition of acid compounds, which explain the pattern with higher historical weathering intensity towards the surface. However, there is an on-going depletion of easily weathered minerals over time in the uppermost part of the soil, which limits the weathering rate towards the surface. The weathering intensity is gradually shifted towards lower depths, where there are more easily weathered minerals present. Since less weathering agents (i.e. organic acids, HCO_3^-) are being consumed in the uppermost part of the soil, they are transported down in the profile and contribute to the weathering at lower depths. In our results, the steady-state weathering rate estimated with PROFILE exceeds the historical rates estimated by the depletion method in deeper soil layers (Fig. 4).

4.2. Applicability of the two approaches for different conditions

The three sites that were outliers in the comparison can be useful to analyze critical conditions for the two approaches. In Nunasvaara the PROFILE weathering rates of Ca, Mg and Na were much higher than for any of the other sites, and also several times higher than the weathering rates estimated with the depletion method. The depletion method indicated moderate losses despite the seemingly good mineralogy indicated by the base cation content. The high modeled weathering rates by PROFILE can be explained by a combination of high contents of minerals rich in Ca, Mg and Na in the soil, e.g. plagioclase, hornblende, biotite and augite (Table 3), high soil density and a high fraction of fine-grained material (Table 1). A possible explanation to the deviation between the methods could be that the simplified handling of hydrology in PROFILE does not take into account that the high soil density may lead to saturation of the soil and thus substantially increased product inhibition, slowing down weathering rates. In Vålberget the situation was the other way around; the depletion method gave many times higher weathering rates for Ca and Mg than PROFILE. The Ca, Na and K weathering rates in Vålberget were the lowest of the 16 sites according to PROFILE and the Mg weathering rate was among the lowest. The depletion method gave by far the highest weathering rates of the 16 sites for Ca and Mg, whereas the K weathering was one of the lowest and the Na weathering was intermediate compared to the other sites. The fractional mass loss of this site was also very large; on average 55% of the Ca and 44% of the Mg has been lost down to 50 cm depth, while in the uppermost layer the result showed that as much as 81% of the Ca and 87% of the Mg has been lost by weathering. An indication that the Ca weathering rate in Vålberget could be problematic was that the site deviated completely from the otherwise strong relationship between Ca and Na weathering estimated by the depletion method. This indicate that the systematic Ca gradient in the profile was caused by heterogeneities rather than soil development and that the weathering rate thus was overestimated. In Målaskog the PROFILE model resulted in much higher weathering rates than the depletion method for Ca and Na. Although the soil in Målaskog was relatively coarse, its high Ca content led to the second highest Ca weathering rate according to PROFILE. The soil profile in Målaskog showed opposite gradients both for mobile and immobile ions indicating vertical heterogeneity in parent material composition, which means that the method assumptions for the depletion method were not fulfilled.

4.3. The results in a sustainability perspective

For detailed sustainability assessments the complete mass balance has to be taken into account, i.e. base cation deposition, weathering, harvest losses and leaching (Sverdrup et al., 2006; Akselsson et al., 2007). Base cation deposition in Sweden is in the same order of magnitude as weathering rates (Akselsson et al., 2007), but the input of base cations through weathering and deposition needs to be substantially higher than the harvest losses, to avoid soil acidification. However, simplified mass balance including only weathering and harvest losses is often used as an indication of the sustainability (Olsson et al., 1993; Klaminder et al., 2011). If weathering rates are substantially higher than base cation removal at whole-tree harvesting, the harvesting effects on the nutrient budget can be expected to be small. However, if the weathering rates are in the same order of magnitude or lower than the base cation removal, there is an obvious risk of negative effects on the soil nutrient status. Our results show that the removal at whole-tree harvesting were in the same order of magnitude or higher level than the weathering rate, no matter which of the approaches that was used. For the sites in southern Sweden, the losses at harvesting were substantially larger than the weathering rates. This area also contains the most acidic soils, due to high historical levels of acid deposition (Pihl Karlsson et al., 2011). Thus, the risk of negative effects associated with base cation losses at harvesting is largest in these regions. The northern sites have better soil status to start with from an acidification point of view, and the losses at whole tree harvesting are smaller. These areas are associated with lower risks, but since the losses at harvesting are as big as the weathering rates, and since the base cation deposition is relatively low in northern Sweden, there is not much left for the runoff water, which may lead to negative effects for surface waters on a long term.

5. Conclusions

Where conditions are suitable for both methods there is a correspondence in weathering rates between the depletion method and PROFILE. For Mg, the depletion method yield higher rates than PROFILE, probably due to strong depletion of main Mg-bearing minerals, such as chlorite and hornblende. For Ca, K and Na the weathering rates are generally higher according to PROFILE than according to the depletion method. This indicates that initial weathering rates of these elements at the time of soil formation were less extreme in the investigated sites compared to other regions. The weathering rates in the upper 50 cm decrease with depth according to the depletion method, but increase with depth according to PROFILE. The pattern can be explained by the methods difference in time perspective; historically most of the weathering has taken place in the upper soil, which is now depleted of easily weathered minerals leading to a lower current rate as represented by PROFILE. The application of multiple methods on a large selection of sites can help identify deviating results for the methods included. The losses from whole-tree harvesting exceed weathering for both the depletion method and PROFILE, especially for sites in southern Sweden.

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Figure captions

Fig. 1. Location of the 16 sampling sites.

Fig. 2. Weathering rate (meq m^{-2} yr^{-1}) for Ca, Mg, K and Na estimated by the PROFILE and the depletion method for 16 soil profiles.

Fig. 3. The fractional mass loss of Ca, Mg, K and Na at different soil depths (cm) estimated by the depletion method. The error bars represent the 95% confidence limits.

Fig. 4. Weathering intensity (meq m^{-3} yr^{-1}) of the sum of base cations vs. soil depth (cm) estimated by the PROFILE and the depletion method. The error bars represent the 95% confidence limits.

Fig. 5. Weathering rates of base cations estimated by the two approaches compared with harvest losses of base cations at eight spruce sites. Harvest losses are calculated for a whole-tree harvesting scenarios where all tree stems, 60% of the branches and 75% of the needles on the branches are harvested. The dashed lines in the harvesting bars indicate the level when only tree stems are harvested.

Tables

Table 1. Site characteristics and soil properties at 50 cm depth in the mineral soil for the 16 investigated sites.

Site	Lat.	Long.	MAT (°C)	MAP (mm)	Clay (%$_{wt}$)	Silt (%$_{wt}$)	Sand (%$_{wt}$)	Density[b] (g dm^{-3})	Soil depth (cm)	Soil age (calendar years)	Soil moisture class[c]	Tree species	Stem volume production (m^3 ha^{-1} yr^{-1})
Bodafors	57.6	14.7	5.4	729	2	38	60	1400	192	14 000	3	Norway spruce	10.7
Gårdsjön	58.0	12.0	6.6	952	12	33[a]	55[a]	1055	72	14 340	3	Norway spruce	9.5
Hjärtasjö	59.0	15.3	5.2	650	5	31[a]	64[a]	992	113	11 280	3	Norway spruce	9.5
Hässlen	60.3	16.3	4.3	629	9	41[a]	50[a]	1442	130	10 830	3	Other broadleaves	8.4
Kloten	59.9	15.3	4.2	807	2	48	50	1590	125	10 910	4	Scots pine	6.1
Kullarna	62.0	16.6	3.8	552	4	12[a]	84[a]	1390	102	10 400	3	Norway spruce	5.2
Lammhult	57.2	14.8	5.5	688	2	35	63	1376	178	14 260	3	Norway spruce	9.7
Målaskog	56.8	14.3	6.4	707	0	23[a]	77[a]	1072	175	14 360	3	Other broadleaves	11.3
Nunasvaara	67.8	21.4	-2.0	456	4	43[a]	53[a]	1852	227	9 880	3	Scots pine	1.7
Risfallet	60.3	16.2	4.3	629	7	35[a]	58[a]	1560	136	10 810	3	Scots pine	7.0
Skånes Värsjö	56.3	13.5	6.4	765	3	25[a]	72[a]	1291	112	14 700	3	Norway spruce	11.6
Stöde	62.3	16.4	2.5	707	2	33[a]	65[a]	1271	263	10 330	3	Norway spruce	5.5
Svartberget	64.3	19.7	1.0	573	2	26[a]	72[a]	1534	217	10 120	4	Norway spruce	3.6
Söderåsen	56.1	13.0	7.2	795	6	24[a]	70[a]	1350	126	15 900	3	Beech	12.6
Vindeln	64.2	19.6	1.2	523	4	37	59	1407	207	10 130	3	Scots pine	3.6
Vålberget	60.8	13.8	2.6	701	3	17[a]	80[a]	1243	176	10 580	3	Clearcut (spruce)	6.2

[a] The fraction 0.02-0.06 mm, which according to common definitions is a part of the silt fraction, was included in sand fraction in the original data for many sites. In this study, the sand and silt fractions were recalculated according to common definitions to be compatible with the PROFILE model. The calculations were based on relationships from the other sites, where complete grain size distributions were available.

[b] Volume weight of < 2 mm fraction including coarser dead organic material (volume of live roots, gravel and stones deducted from sampled volume).

[c] Drainage class according to the NORDSOIL database: 1. Excessively, 2. Somewhat excessively, 3. Well, 4. Moderately, 5. Imperfectly, 6. Poorly 7. Very poorly.

Table 2. Element concentrations for the < 2 mm soil fraction at the reference layer depth used in the depletion method estimates.

Site	Ref. depth (cm)	Ca (%)	Mg (%)	K (%)	Na (%)	Zr (ppm)
Bodafors	62	1.47	0.65	2.31	2.03	298
Gårdsjön	57	1.60	0.68	2.54	2.29	299
Hjärtasjö	57	1.13	0.88	1.99	1.62	189
Hässlen	82.5	1.19	0.79	3.10	1.72	250
Kloten	48.5	0.94	0.28	2.47	1.76	247
Kullarna	62	1.14	0.58	2.99	1.84	343
Lammhult	98	1.54	0.59	2.34	2.31	232
Målaskog	60.0	2.39	1.07	2.47	2.66	261
Nunasvaara	54.5	2.95	2.04	1.88	3.29	192
Risfallet	76	1.01	0.61	2.55	2.05	181
Skånes Värsjö	62	1.00	0.24	3.10	2.33	466
Stöde	55.5	1.47	0.86	2.32	1.87	229
Svartberget	154	1.37	0.41	2.41	2.35	241
Söderåsen	101	0.75	0.36	2.84	1.8	341
Vindeln	129	1.00	0.92	2.47	1.67	279
Vålberget	117	1.57	0.77	1.36	0.79	186

Table 3. Normative mineralogy at 50 cm depth in the mineral soil based on the A2M model (Qu=Quartz, Or=Orthoclase, Pl=Plagioclase, Ho=Hornblende, Ep=Epidote, Bi=Biotite, Au=Augite, Ap=Apatite, Mu=Muscovite, Ch=Chlorite, Il=Illite, Ve=Vermiculite, An/Ab=ratio between Anorthite and Albite in Plagioclase).

	Mineral contents (%)												
	Qu	Or	Pl	Ho	Ep	Bi	Au	Ap	Mu	Ch	Il	Ve	An/Ab
Bodafors	41.0	13.2	25.8	1.2	3.8	0.0	0.0	0.5	3.6	2.5	1.6	5.5	0.12
Gårdsjön	36.5	4.7	26.4	1.0	3.6	0.0	0.0	0.2	8.4	1.4	12.2	3.5	0.14
Hjärtasjö	49.3	3.0	20.4	0.8	1.9	0.0	0.0	0.2	9.4	1.9	9.1	4.1	0.12
Hässlen	40.2	15.1	21.8	1.0	2.5	0.0	0.0	0.2	6.3	2.1	6.6	3.6	0.13
Kloten	51.5	13.6	21.8	0.5	3.0	0.0	0.0	0.2	3.4	1.0	3.0	1.3	0.10
Kullarna	39.1	15.3	25.6	1.0	2.9	0.0	0.0	0.2	5.5	1.8	4.8	2.5	0.15
Lammhult	37.9	14.6	29.0	1.5	4.2	0.0	0.0	0.4	2.4	2.0	1.2	4.9	0.12
Målaskog	28.5	15.7	31.8	2.6	7.5	0.0	0.0	0.6	2.6	2.7	1.3	5.2	0.11
Nunasvaara	16.7	10.2	36.0	11.1	2.1	2.7	8.1	0.5	0.0	1.3	4.5	4.7	0.05
Risfallet	45.4	13.5	26.2	0.9	2.6	0.0	0.0	0.2	3.8	2.1	3.4	3.2	0.10
Skånes Värsjö	38.8	16.5	29.7	0.7	2.4	0.0	0.0	0.3	4.3	1.0	2.2	1.4	0.10
Stöde	40.5	4.7	23.4	1.9	3.7	0.0	0.0	0.4	9.9	3.6	9.3	2.5	0.13
Svartberget	42.4	15.1	29.1	1.2	2.9	1.6	0.5	0.2	0.0	0.8	4.5	1.9	0.15
Söderåsen	48.9	10.2	21.3	0.5	1.4	0.0	0.0	0.3	9.9	0.7	4.9	0.9	0.08
Vindeln	39.1	9.8	24.3	1.0	2.3	1.2	0.5	0.3	0.0	0.7	17.1	5.4	0.09
Vålberget	66.3	6.4	8.9	2.6	2.5	0.0	0.0	0.3	3.6	1.6	0.8	5.2	0.22

Table 4. Spearman rank correlation coefficients between weathering estimates by PROFILE and the depletion method where ∑ indicates the sum of base cations (n=16, p-values in italic).

		Depletion method				
		Ca	Mg	K	Na	∑
PROFILE	Ca	0.44	0.20	0.31	0.59	0.36
		0.09	*0.46*	*0.24*	*0.02*	*0.16*
	Mg	0.12	0.51	0.23	0.22	0.24
		0.65	*0.04*	*0.40*	*0.42*	*0.36*
	K	-0.13	-0.01	0.07	0.18	-0.07
		0.63	*0.97*	*0.80*	*0.50*	*0.80*
	Na	0.05	0.07	0.15	0.25	0.04
		0.85	*0.80*	*0.57*	*0.34*	*0.88*
	∑	0.19	0.11	0.14	0.34	0.11
		0.49	*0.69*	*0.62*	*0.19*	*0.67*

Figures

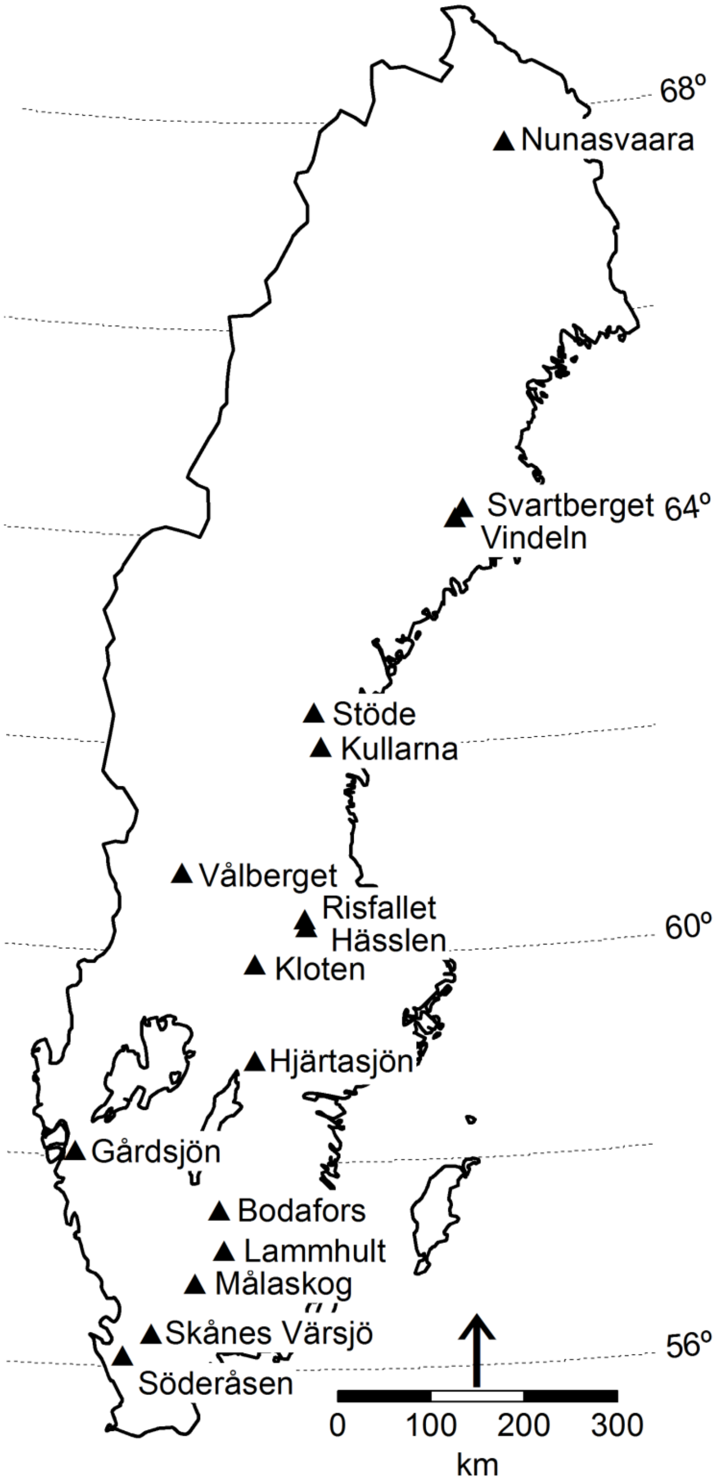
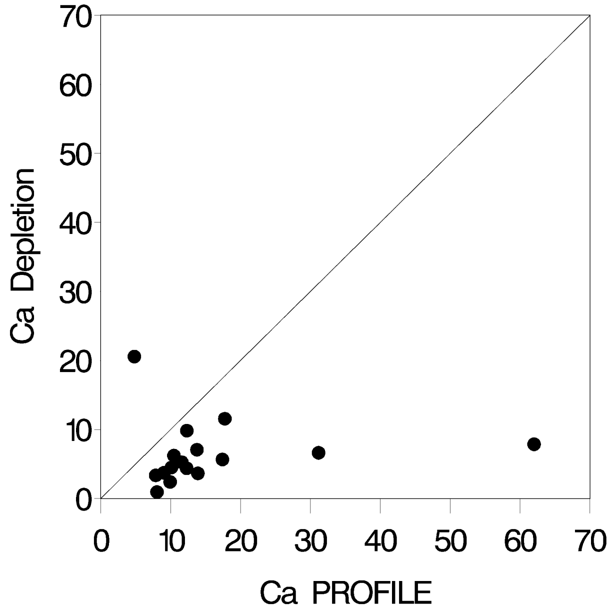


Fig.1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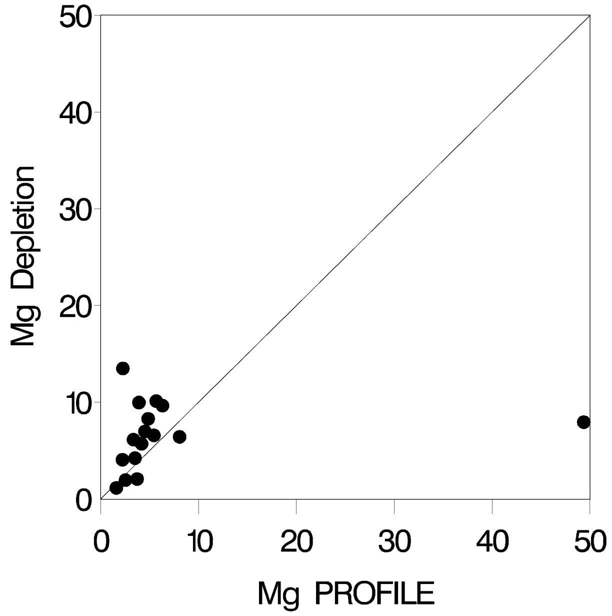
0
10
20
30
40
50
60
70

Fig. 2a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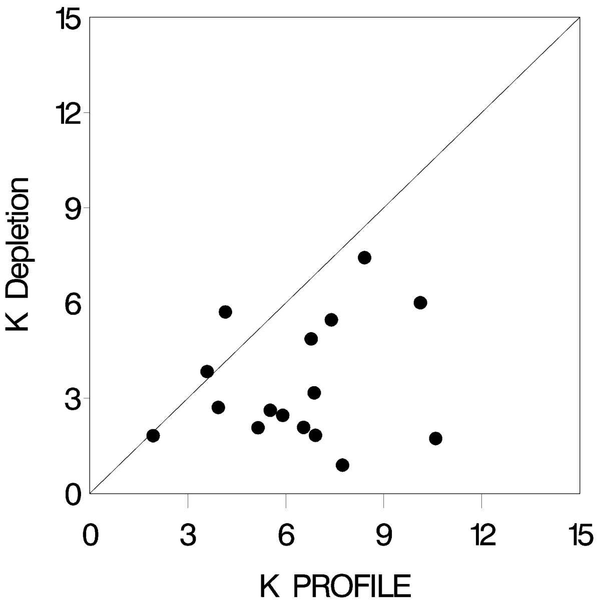
10
20
30
40
50

Fig. 2b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15
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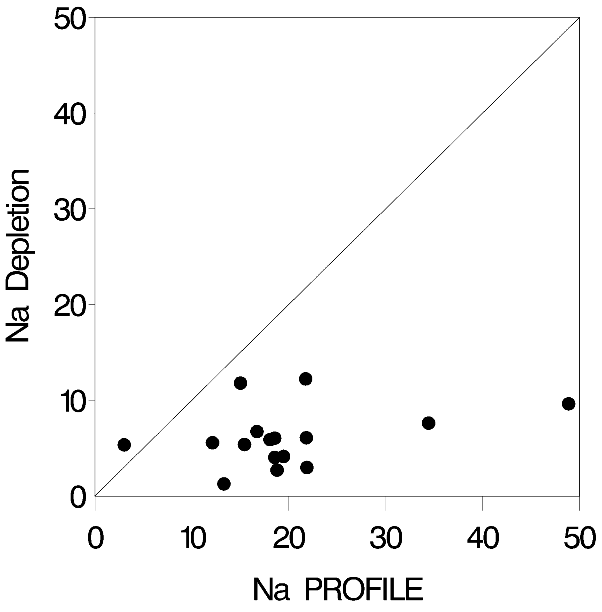
12
15

Fig. 2c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50
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40
50

Fig. 2d

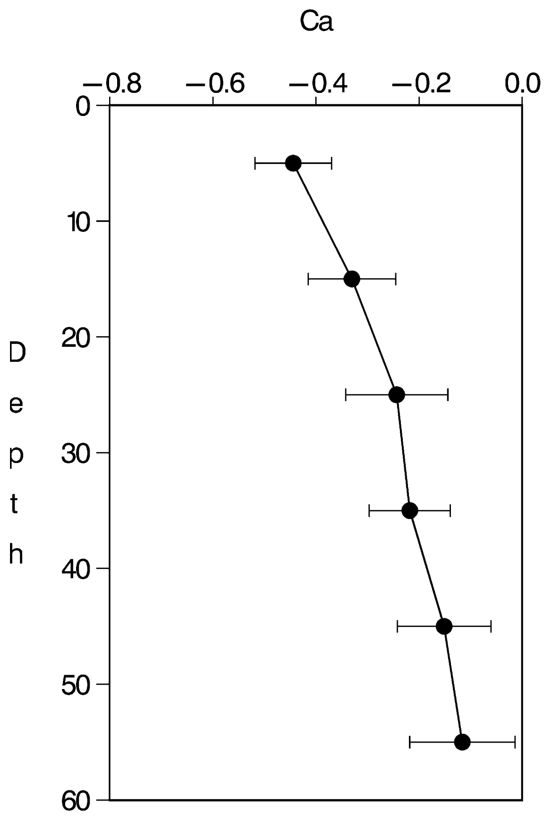
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Fig. 3a

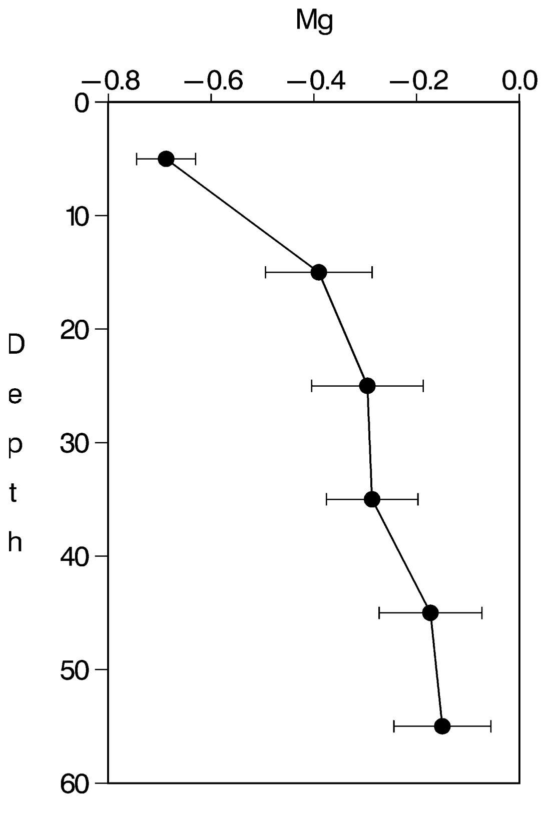
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Fig. 3b

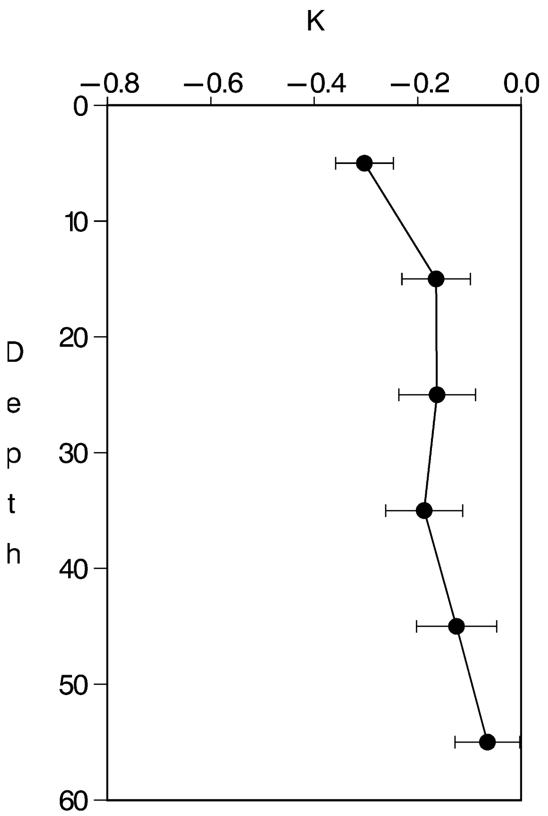
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Fig. 3c

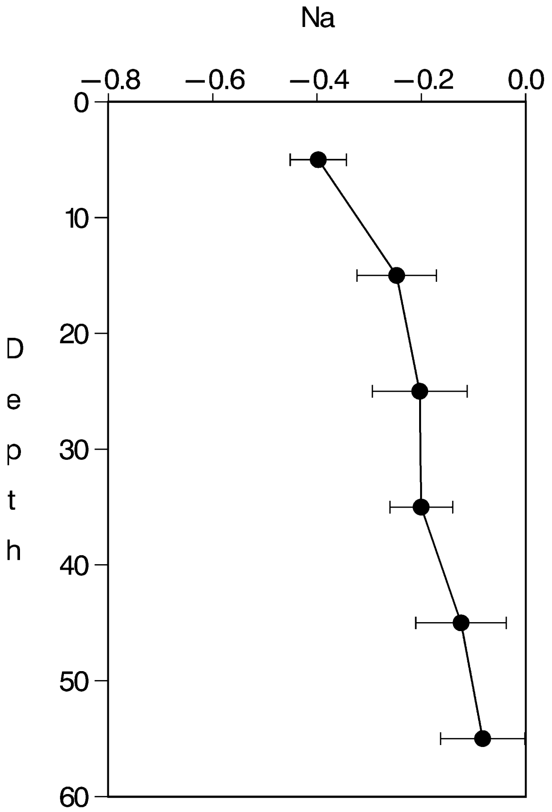
Na
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Fig. 3d

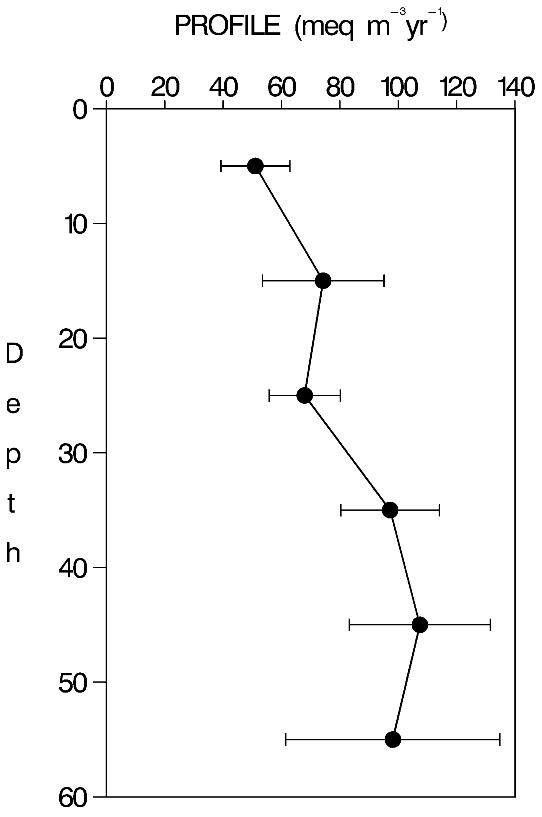

PROFILE (meq $m^{-3}yr^{-1}$)
0
20
40
60
80
100
120
140
0
10
20
30
40
50
60
Depth

Fig. 4a

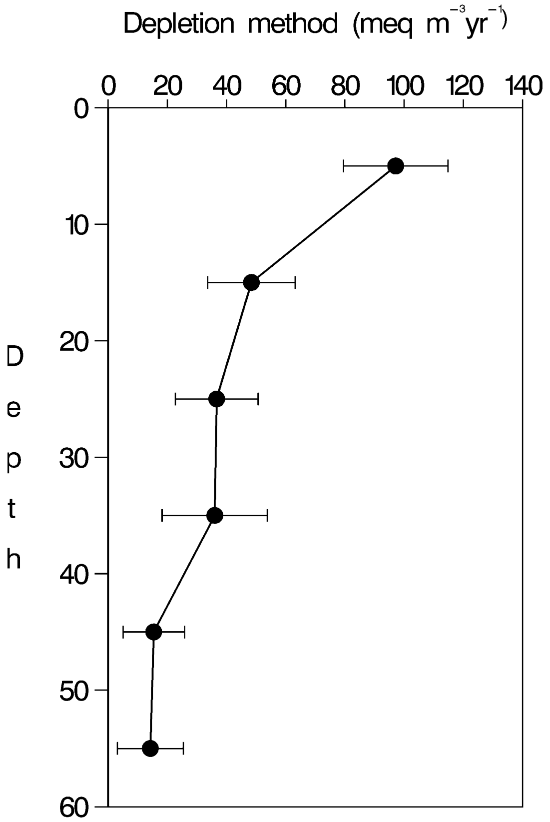
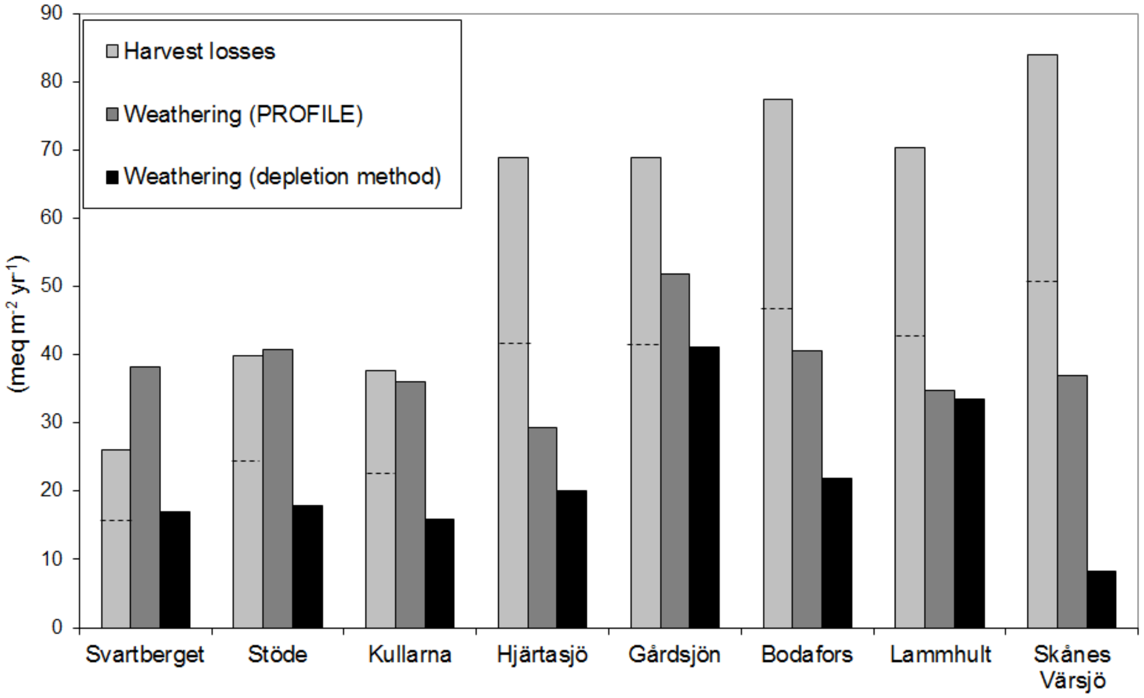

Depletion method (meq $m^{-3}yr^{-1}$)
0
20
40
60
80
100
120
140
0
10
20
30
40
50
60
Depth

Fig. 4b



90
80
70
60
50
40
30
20
10
0
(meq m-2 yr-1)
Harvest losses
Weathering (PROFILE)
Weathering (depletion method)
Svartberget
Stöde
Kullarna
Hjärtasjö
Gårdsjön
Bodafors
Lammhult
Skånes Värsjö

Fig. 5